

**(19) World Intellectual Property Organization  
International Bureau**



(10) International Publication Number  
**WO 03/035241 A1**

(43) International Publication Date  
1 May 2003 (01.05.2003)

PCT

(51) **International Patent Classification<sup>7</sup>:** B01J 8/02,  
F28F 3/14, F28D 9/00

(21) **International Application Number:** PCT/EP02/11024

(22) **International Filing Date:** 2 October 2002 (02.10.2002)

(25) **Filing Language:** English

(26) **Publication Language:** English

(30) **Priority Data:**  
01124956.2 19 October 2001 (19.10.2001) EP

(71) **Applicant (for all designated States except US):** METHANOL CASALE S.A. [CH/CH]; Via San Carlo 22, CH-6932 Breganzona (CH).

(72) **Inventors; and**

(75) **Inventors/Applicants (for US only):** FILIPPI, Ermanno [IT/CH]; Strada di Gandria 73, CH-6976 Castagnola (CH).

(81) **Designated States (national):** AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, OM, PH, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.

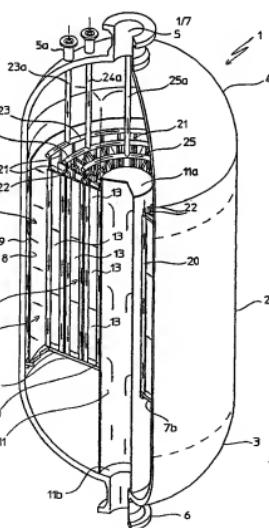
(84) **Designated States (regional):** ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TI, TM), European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE,

[Continued on next page]

**(54) Title: HEAT EXCHANGE UNIT FOR ISOTHERMAL CHEMICAL REACTORS**



WO 03/035241 A1



**(57) Abstract:** A heat exchange unit for isothermal radial or axial-radial chemical reactors (1), with an overall configuration which is substantially annular and cylindrical having a passage (11) with a predetermined diameter running through it axially, is distinguished in that it comprises a plurality of substantially rectangular, flattened box-shaped heat exchangers (13), distributed in many coaxial and concentric arrays, defining in the unit (12) a plurality of radial alignments with a substantially radial arrangement, and wherein the exchangers (13) have long sides (13a) parallel to the axis of the unit (12) and short sides (13b) extending radially, each of the exchangers (13) comprising an inner chamber (17), intended to be passed through by an operating heat exchange fluid, at least one operating pipe (19) and at least one collector pipe (20) of the operating fluid, associated with two respective opposing sides (13a) of the exchanger (13) and extending along them, the pipes (19, 20) being in fluid communication, on the one side, with the chamber (17) through at least one opening (19a, 20a) formed therein and, on the other side, with the outside of the exchanger (13), through respective inlet and outlet fittings (21, 22) for the operating fluid.



ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

*For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

**Published:**

— *with international search report*

Title: "Heat exchange unit for isothermal chemical reactors.

古文 古文 古文 古文

#### DESCRIPTION

## 5 Field of application

The present invention relates, in its broader aspect, to a chemical reactor of the type comprising a substantially cylindrical shell (or pressure vessel) closed at its opposite ends by respective bottoms, a reaction zone in said shell wherein at least a catalytic bed is placed, and a heat exchange unit embedded in said catalytic bed.

Such a reactor is particularly useful for carrying out exothermic and endothermic reactions occurring in substantially isothermal conditions, i.e. conditions in which the reaction temperature is controlled in a rather limited range of values around a predetermined value.

In the following description and claims, a reactor of the aforementioned type is referred to as pseudoisothermal or, in short, isothermal reactor.

20 More in detail, the present invention relates to an  
isothermal reactor of the type being considered, structured  
internally to allow the reactant gases and the gaseous  
reaction products to cross the catalyst bed in a radial or  
axial-radial direction (with reference to the axis of the  
25 cylindrical shell of said reactor) and comprising a heat  
exchange unit made of a plurality of plate-shaped heat  
exchangers, which are substantially box-like, defining in  
their interior a chamber intended to be crossed by a heat  
exchange operating fluid.

30 Prior art

Generally speaking, the operating fluid used in the heat

- 2 -

exchange unit of an isothermal reactor, can be constituted of a fluid whose temperature changes (a non-boiling fluid), such as for example an appropriate and controlled flow of the reactant gases fed to the reactor itself; or a boiling, 5 i.e. isothermal, fluid, such as boiling water, melted salts, Dow Therm, and alike. Furthermore, in order to make the heat exchange more effective, and hence, of improving the kinetics of the chemical reaction, the flow of said operating fluid is directed, inside the respective heat 10 exchange unit, in countercurrent or in co-current with the gaseous reaction products that, in turn, cross the catalyst bed in a radial or axial-radial direction, with reference to the generally vertical axis of the shell of the reactor considered. When the operating fluid consists of the 15 reactant gases themselves, besides removing or providing heat to the reaction environment (catalytic bed) so as to promote the completion of the reaction itself, they warm up or cool down, respectively, reaching a temperature condition which is thermodynamically favourable for the 20 start of the reaction.

In this case, (radial or axial-radial flow) the general configuration of an isothermal reactor is that of a pressure vessel, wherein is supported at least one catalytic cartridge, enclosed by two walls which are 25 concentric and coaxial to the vessel itself and which are suitably perforated to allow the gases to pass through the catalyst contained in said cartridge (catalytic bed). In the mass of catalyst is embedded a heat exchange unit, made of a plurality of exchangers, which are appropriately 30 structured, supported and interconnected.

Amongst the most efficient and most recently used exchangers in isothermal reactors of the type considered here, plate exchangers that have a substantially flattened box-like structure are the most popular.

35 As much as they are advantageous under different points of

view, as for example that of the large surface made available for the heat exchange, the plate-shaped heat exchangers of the aforementioned type suffer from a technical drawback consisting of the difficulty in 5 directing the flow of operating fluid in their interior in the designated direction, for example and in particular in the radial direction with respect to the axis of the reactor. This is a difficulty, which often means a reduced heat exchange efficiency and, therefore, a reduced yield of 10 the chemical reaction that is intended to be developed.

Summary of the invention

The technical problem underlying the present invention is that of providing a heat exchange unit for radial or axial-radial isothermal reactors, having structural and 15 functional characteristics such as to overcome the drawback quoted above with reference to the prior art.

This problem is solved according to the invention by a heat exchange unit for isothermal radial or axial-radial chemical reactors, with an overall configuration which is 20 substantially annular and cylindrical having a passage with a predetermined diameter running through it axially, characterized in that it comprises a plurality of substantially rectangular, flattened box-shaped heat exchangers, distributed in many coaxial and concentric 25 arrays, defining in said unit a plurality of radial alignments with a substantially radial arrangement, and wherein said exchangers have long sides parallel to the axis of said unit and short sides extending radially; each of said exchangers comprising an inner chamber, intended to 30 be passed through by an operating heat exchange fluid, at least one distributor pipe and at least one collector pipe of said operating fluid, associated with two respective opposing sides of said exchanger and extending along them, said pipes being in fluid communication, on the one side, 35 with said chamber through at least one opening formed

- 4 -

therein and, on the other side, with the outside of said exchanger, through respective inlet and outlet fittings for said operating fluid.

Brief description of the drawings

5 Fig. 1 schematically represents, in a partial section perspective view, a radial isothermal reactor, using a heat exchange unit comprising a plurality of exchangers according to the invention;

10 Fig. 2 represents on an enlarged scale a heat exchanger of the heat exchange unit of fig. 1;

Figs. 3, 4 and 5 are section views along lines III-III, IV-IV and V-V of fig. 2, respectively;

Fig. 6 schematically represents a preferred configuration of the heat exchanger of fig. 2,

15 Fig. 7 represents a section view along line VII-VII of fig. 6,

Fig. 8 represents an alternative embodiment of the heat exchanger of fig. 6,

20 Fig. 9 represents on an enlarged scale a section along line IX-IX of fig. 8,

Fig. 10 represents an alternative embodiment of the radial isothermal reactor of figure 1.

Detailed description

25 With reference to figure 1, with 1 is wholly indicated an isothermal reactor comprising a cylindrical shell 2, with a vertical axis, closed at the opposite ends with respective bottoms, lower 3 and upper 4, conventionally equipped with passages 5, 6 for the introduction and the discharge of the reactant gases and of the gaseous reaction products into

- 5 -

and from said isothermal reactor 1, respectively, as will become clearer from the rest of the description.

In the shell 2 is defined a reaction zone, in which is conventionally supported a catalytic basket 7, with a substantially cylindrical configuration, having an annular transversal section. Said basket 7 substantially consists of an outer cylindrical wall 8, which, together with said shell 2, defines a space 9 with a reduced width, an inner cylindrical wall 10 and an annular bottom of the conventional type outlined in 7b.

The inner wall 10 centrally defines an axial passage, in which is generally supported a pipe 11, for collecting gaseous reaction products, which has a closed upper end 11a and an open lower end 11b which is in direct fluid communication with the passage 6 of the bottom 3.

Said walls, outer 8 and inner 10, are perforated to allow the passage of the reactant gases from the airspace 9 inside said basket 7, and the passage of the gaseous reaction products from said basket 7, to said central pipe 11.

The basket 7 is intended to contain a mass of a suitable catalyst (not represented), in which is embedded and supported, in a *per se* known way, a heat exchange unit globally indicated with 12.

Said heat exchange unit 12 has an overall annular cylindrical configuration, similar to that of said basket 7. It has an outer diameter that is substantially equal to the outer diameter of the basket 7 and is axially crossed by a passage having a diameter substantially equal to the inner diameter of the basket itself.

In particular, according to a preferred but not limiting embodiment, outlined in Fig. 1, said heat exchange unit 12 comprises a plurality of heat exchangers 13, regularly

- 6 -

distributed in three coaxial and concentric arrays. Each exchanger 13 (fig. 2) has a substantially box-like structure, with an essentially elongated, flattened rectangle configuration, in which two opposite long sides 13a and two opposite short sides 13b are outlined.

5 In the aforementioned heat exchange unit 12, the exchangers 13 are in a substantially radial arrangement, with long sides 13a parallel to the axis of said unit 12 (and therefore to the axis of the shell 2 of said reactor 1), 10 and short sides 13b extending radially, and are organised to constitute a plurality of radial sets of three exchangers.

15 Of course, according to different technical and implementing requirements, each set of three exchangers 13 can be replaced with a pair of exchangers or by one single exchanger, which occupies the entire radial extension of the basket 7.

20 Even more particularly (figs. from 2 to 5) each exchanger 13 is made of a pair of juxtaposed metallic sheets 14, 15, mutually joined at a predetermined distance by perimetric 25 soldering 16, 16a, so that between them a chamber 17 is defined, intended to be crossed by an operating heat exchange fluid.

25 For an optimal heat exchange efficiency with the reactant gases and with the gaseous reaction products which cross the catalytic bed in the radial direction, the heat exchange fluid must in turn cross each exchanger 13 of the exchange unit 12 in a radial or substantially radial direction, in co-current or in countercurrent to said 30 gases.

For such a purpose and in accordance with a characteristic of the present invention, each exchanger 13 is equipped at an opposite long sides 13a, with a distribution pipe 19 and a

collection pipe 20, respectively, for said operating fluid. The pipes 19 and 20 are, on one side, in fluid communication with said chamber 17 through at least one, but preferably a plurality of openings or holes 19a and 20a, with which they are equipped along one or more generatrix and, on the other side, with the outside of the exchanger 13, through fittings 21 and 22, for the inlet and outlet of said operating fluid, respectively.

According to a first preferred embodiment, said pipes 19 and 20 are "formed" directly in the long sides 13a of the exchanger 13, at the moment of the drawing and perimetric soldering of the metallic sheets 14 and 15 which make it up. Advantageously, they are obtained through welding seams 116, 116a, extending parallel to the perimetric soldering 16 and 16a, at a predetermined distance from them, whereas the openings 19a, 20a for the passage of fluid are obtained through appropriate interruptions of such welding seams 116, 116a.

According to another embodiment (not represented) said pipes 19 and 20 are made of respective tubes, fixed in said chamber 17, at the long sides 13a, of the exchanger and parallel to them and form a whole with the respective fittings 21 and 22, quoted above.

In accordance with another characteristic of the present invention, the fittings 21 and 22 of the pipes 19 and 20, for distributing and collecting the operating fluid into and from each exchanger 13, respectively, are arranged at a same short side 13b of the exchanger.

When arranged to constitute said heat exchange unit 12 of the present invention, in the arrangement described above (Fig. 1), the short side 13b, with corresponding fittings 21 and 22, constitutes the upper side of every exchanger 13, whereas the opposite short side is directly fixed, in a *per se* known way, to a support structure (not represented)

- 8 -

of the entire heat exchange unit 12.

Advantageously, for greater heat exchange efficiency, at least part of the exchangers 13, of the entire exchange unit 12, are manufactured according to the configuration 5 schematically illustrated in fig. 6.

According to this alternative embodiment, the inner chamber of each exchanger 13 is subdivided into a plurality of chambers 117, not directly communicating with each other and obtained, for example, through a corresponding 10 plurality of welding seams 117a of the metallic sheets 14, 15, extending parallel to the short sides 13b of the exchanger 13, in other words perpendicular to its distributor 19 and collector 20 pipes. Said chambers 117, which can either all have the same width or have different 15 widths depending on requirements, are internally equipped with a plurality of deflector baffles 118, extending parallel to said pipes 19, 20 and which define in each chamber 117, a substantially serpentine fluid path.

Each chamber 117 is in fluid communication with said 20 distributor pipe 19, through at least one opening 19a thereof and with said collector pipe 20, through at least one opening 20a thereof.

It should be noted that, for a better control of the pressure drop, and therefore of the distribution of the 25 operating fluid inside the chambers 117, the openings 19a of the distributor pipe 19, are provided near the bottom of each of said chambers 117, and with a different width or light, in particular with an increasing width in the flow direction of the operating fluid inside said pipe 19.

30 Advantageously, to increase the aforementioned possibility of controlling the distribution of the operating fluid in the chambers 117, when the pressure drop therein is too low to guarantee it, the use of calibrated sleeves 119a (figs.

8 and 9) are foreseen, appropriately inserted in each of said openings 19a, so as to obtain precise and calculated fluid-passage sections (and thus precise and calculated pressure drop).

- 5 In the isothermal reactor 1 of the present invention, at the upper end of the catalytic basket 7 and in a position overlying the heat exchange unit 12, an annular pipe 23 is foreseen for the general distribution of the operating fluid.
- 10 When the operating fluid consists of the reactant gases themselves (fig. 1), and must cross the heat exchange unit 12 in co-current with the gaseous reaction products, such a pipe 23 is in fluid communication with the outside of the isothermal reactor 1, through a respective feeding pipe 23a 15 of said fluid, connected to an inlet passage 5a of said reactant gases in the reactor itself. On the other side, said pipe 23 is in fluid communication with the plurality of distributor pipes 19, of the outermost exchangers 13 in each radial set of three exchangers.
- 20 Moreover, in each of said radial sets of three, the collector pipes 20 of each exchanger are directly connected with the distributor pipes 19 of the exchanger which immediately follows it towards the axis of the shell 2. Such a connection can be carried out by putting the 25 collector fittings 22 of an exchanger in fluid communication with the distributor fittings 21 of the subsequent exchanger, in a sort of head-to-tail hydraulic connection, as shown in the attached drawings.

The collector pipes 20 of the more inner exchangers 13 of 30 the heat exchange unit 12, are "open" in the upper part of the shell 2 (substantially at the upper bottom 4), so that the reactant gases preheated or cooled in the exchangers 13 can be conveyed to the space 9 and from there to the catalytic bed contained in the basket 17 which is crossed

- 10 -

in a radial direction.

With the arrangement described above it is possible to achieve a further important advantage. Indeed, in the same zone occupied by the annular distributor pipe 23, it is possible to arrange another two (or more) annular pipes 24, 25, for the distribution of the predetermined operating heat exchange fluid (in particular and advantageously consisting of reactant gases), each of which is in fluid communication with the heat exchangers 13 of a respective annular array of exchangers. For example and preferably, such a connection is provided at the head-to-tail hydraulic connections between exchangers 13 of a same radial set of three exchangers.

With 24a and 25a are indicated feeding pipes of said operating fluid from the outside of the reactor to the annular pipes 24 and 25, respectively.

In this way, it is possible to introduce "fresh" operating fluid into the exchangers 13 of the intermediate annular arrays and therefore it is possible to control the temperature profile in the catalytic bed.

In fig. 10 an alternative embodiment of the isothermal reactor of the present invention is represented, particularly, even if not exclusively, suitable when the operating fluid to be used is water, or else reactant gases to be preheated (or cooled), but to be made to react in another reactor. In this figure, all of the details already described in fig. 1 are defined with the same reference numbers.

According to this alternative, in the zone which is (possibly) occupied by the annular pipes 23, 24 and 25, for the distribution of the operating heat exchange fluid, a final annular pipe 26 (collector pipe), is arranged to collect said operating fluid which is discharged from the

- 11 -

heat exchange unit of the present invention. For such a purpose, the annular pipe 26 is in fluid communication, on one side, with all of the collector pipes 20 through the fitting 22 of the innermost heat exchangers 13 of said unit 12 and, on the other side, through a pipe 26a, with an "outlet" nozzle 27 from the isothermal reactor. The reactant gases are introduced into the reactor 1 through the upper passage 5, provided therein.

For the purpose of discharging the catalyst without removing the plate-shaped exchangers 13, the basket 7 of the chemical reactor of the present invention is equipped with a bottom wall 28, which is convex towards the inside of the basket itself. In turn, this bottom wall 28 is equipped, near to the outer wall of said basket 7, with a plurality of openings 29 (in fig. 10 only one opening can be seen), in communication with respective nozzles 30 for the discharge of catalyst, provided in the bottom 3 of said reactor.

The invention thus conceived is susceptible to further variants and modifications all falling within the skill of the man skilled in the art and, as such, falling within the scope of protection of the invention itself, as it is defined by the following claims.

## CLAIMS

1. Heat exchange unit for isothermal radial or axial-radial chemical reactors (1), with an overall configuration which is substantially annular and cylindrical having a passage 5 (11) with a predetermined diameter running through it axially, characterized in that it comprises a plurality of substantially rectangular, flattened box-shaped heat exchangers (13), distributed in many coaxial and concentric arrays, defining in said unit (12) a plurality of radial alignments with a substantially radial arrangement, and wherein said exchangers (13) have long sides (13a) parallel to the axis of said unit (12) and short sides (13b) extending radially, each of said exchangers (13) comprising an inner chamber (17), intended to be passed through by an 10 operating heat exchange fluid, at least one distributor pipe (19) and at least one collector pipe (20) of said operating fluid, associated with two respective opposing sides (13a) of said exchanger and extending along them, said pipes (19, 20) being in fluid communication, on the one side, with said chamber (17) through at least one opening (19a, 20a) formed therein and, on the other side, with the outside of said exchanger (13), through respective 15 inlet and outlet fittings (21, 22) for said operating fluid.
2. Heat exchange unit according to claim 1, characterized in that each of said exchanger (13) is made of a pair of juxtaposed metallic sheets (14, 15), mutually joined at a predetermined distance by perimetric soldering (16) so that between them said chamber (17) is defined.
3. Heat exchange unit according to claim 2, characterized in that said pipes (19, 20) for distributing and collecting the operating fluid, respectively, are arranged at opposed 20 long sides (13a) of a respective exchanger (13)..
4. Heat exchange unit according to claim 3, characterized

- 13 -

in that said pipes (19, 20) consist of respective tubes, fixed in said chamber (17) at said opposite long sides (13a) of the respective exchanger (13).

5. Heat exchange unit according to claim 3, characterized in that said pipes (19, 20) are directly formed at said long sides (13a) while forming the exchanger.

6. Heat exchange unit according to claims 4 and 5, characterized in that said pipes (19, 20) are in fluid communication with the outside of the exchanger (13) through respective tubular fittings (21, 22), placed on a same short side (13b) of the exchanger itself and facing the same part of said unit (12).

7. Heat exchange unit according to claims 4 and 5, characterized in that said pipes (19, 20) are in fluid communication with said chamber (17) through respective plurality of openings (19a, 20a), formed in said pipes (19, 20) along generatrix thereof.

8. Heat exchange unit according to claim 1, characterized in that in said radial arrangement and in each radial alignment of heat exchangers (13), each exchanger (13) has the said respective fitting (22) for the outlet of the operating fluid, in fluid communication with the fitting (21) for the fluid inlet of the exchanger (13) adjacent in the same radial alignment, through a substantially head-to-tail hydraulic connection.

9. Heat exchange unit according to claim 1, characterized in that said chamber (17) is subdivided in a plurality of chambers (117) that do not communicate with each other directly, each of which is in fluid communication with said distribution pipe (19) and with said collector pipe (20), through respective openings (19a, 20a) formed therein.

10. Heat exchange unit according to claim 9, characterized in that said chambers (117) are obtained through welding

- 14 -

seams (177a) of said metallic sheets (14, 15) extending perpendicularly to said pipes (19, 20).

11. Heat exchange unit according to claim 10, characterized in that each of said chambers (117) is internally equipped with a plurality of deflector baffles (118), extending parallel to said pipes (19, 20) and which define a substantially serpentine path for said operating fluid.

5 12. Heat exchange unit according to claim 11, characterized in that said chambers have the same width.

10 13. Heat exchange unit according to claim 7, characterized in that said openings (19a) of said distribution pipe (19) are of different width.

15 14. Heat exchange unit according to claim 12, characterized in that said openings (19a) are of increasing width or light in the direction of the flow of said operating fluid inside said distribution pipe (19).

15 15. Heat exchange unit according to claim 12, characterized in that in each of said openings (19a) a calibrated sleeve (119a) is fitted, in order to obtain fluid-passage sections of predetermined dimensions between said distribution pipe (19) and each of said chambers (117).

20 16. Heat exchange unit according to claim 1, characterized in that it comprises at least an annular pipe (23) for the general distribution of said operating fluid, in communication, on one side, with the distribution pipes (19) of the outermost exchangers (13) of said unit (12) and, on the other side, with a respective pipe (23a) for feeding the operating fluid into said reactor (1).

30 17. Heat exchange unit according to claims 8, 16, characterized in that it comprises at least a further annular pipe (24, 25) for the inlet of operating fluid, in communication, on one side, with the head-to-tail hydraulic

- 15 -

connections of each radial alignment of exchangers (13) and, on the other side, with a respective pipe (24a, 25a) for feeding said operating fluid into said reactor (1).

18. Heat exchange unit according to claim 17, characterized in that said collecting pipes (20) of the innermost heat exchangers (13) of said unit (12) are singularly open towards the outside of said unit and at a same side thereof.

19. Heat exchange unit according to claim 18, characterized in that it comprises a further annular pipe (26) for the general collection of the operating fluid discharged from said unit (12), which is connected to all said collecting pipes (20) of the innermost heat exchangers (13) of said unit (12).

15 20. Isothermal chemical reactor comprising a cylindrical shell (2), a catalytic basket (7), supported in said shell (2) and having cylindrical and coaxial outer and inner walls (8, 10), respectively, and a bottom wall (7b, 28), characterized in that it comprises a heat exchange unit (12) according to any claim from 15 to 20, supported in said basket (7).

21. Reactor according to claim 22, characterized in that said bottom wall (28) is convex towards the inside of said basket (7).

25 22. Reactor according to claim 23, characterized in that said convex bottom (28) is equipped with at least one opening (29) for the discharge of catalyst, formed in proximity of the outer wall (8) of said basket (7) and in communication with the outside of said reactor (1).

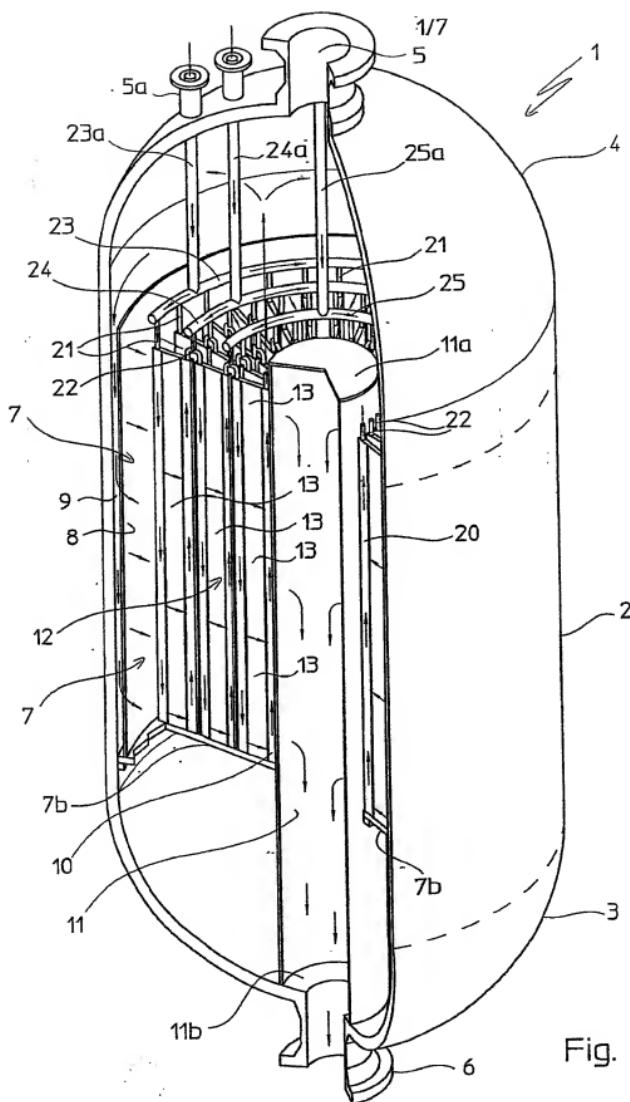


Fig. 1

2/7

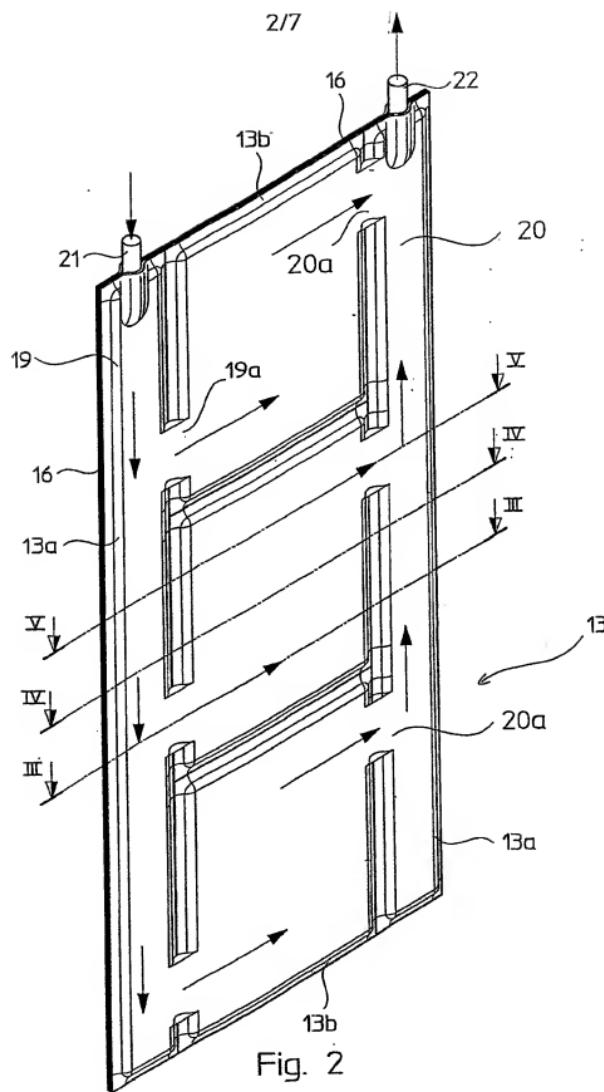


Fig. 2

3/7

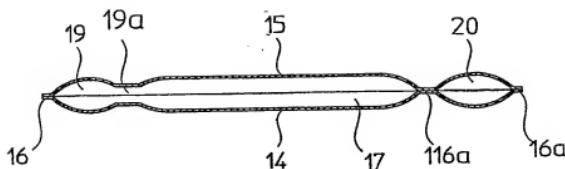


Fig. 3

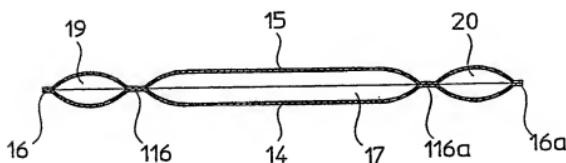


Fig. 4

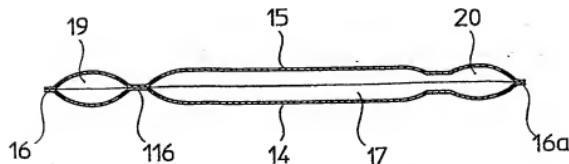
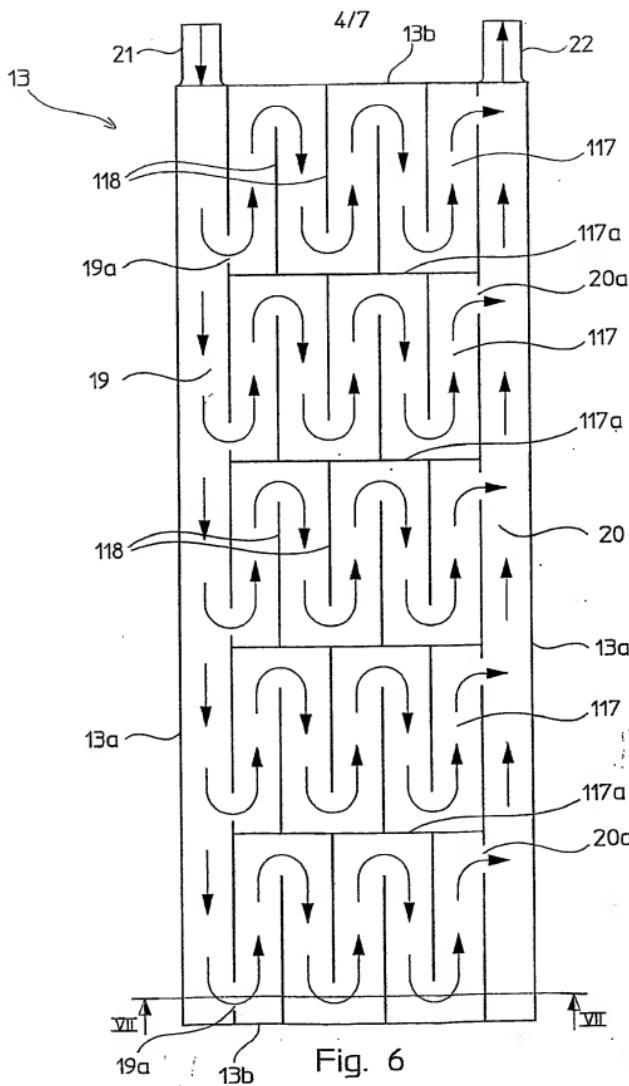


Fig. 5



5/7

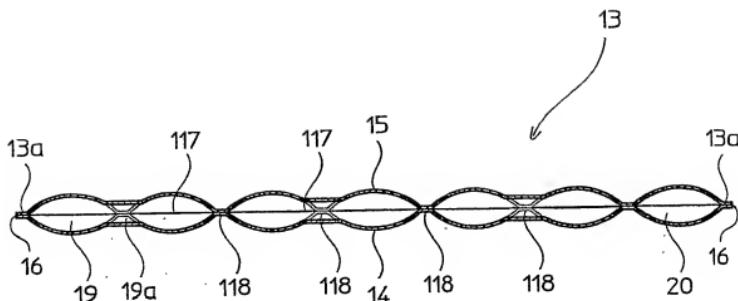


Fig. 7

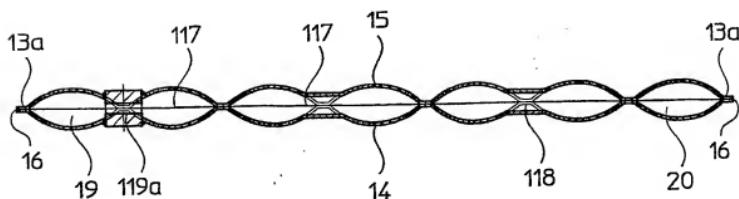


Fig. 9

6/7

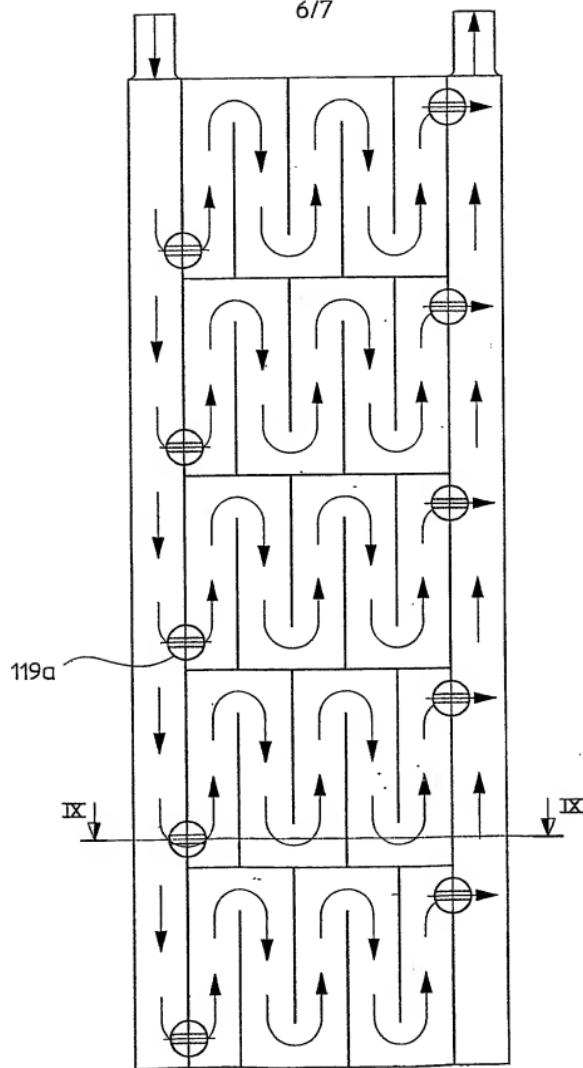
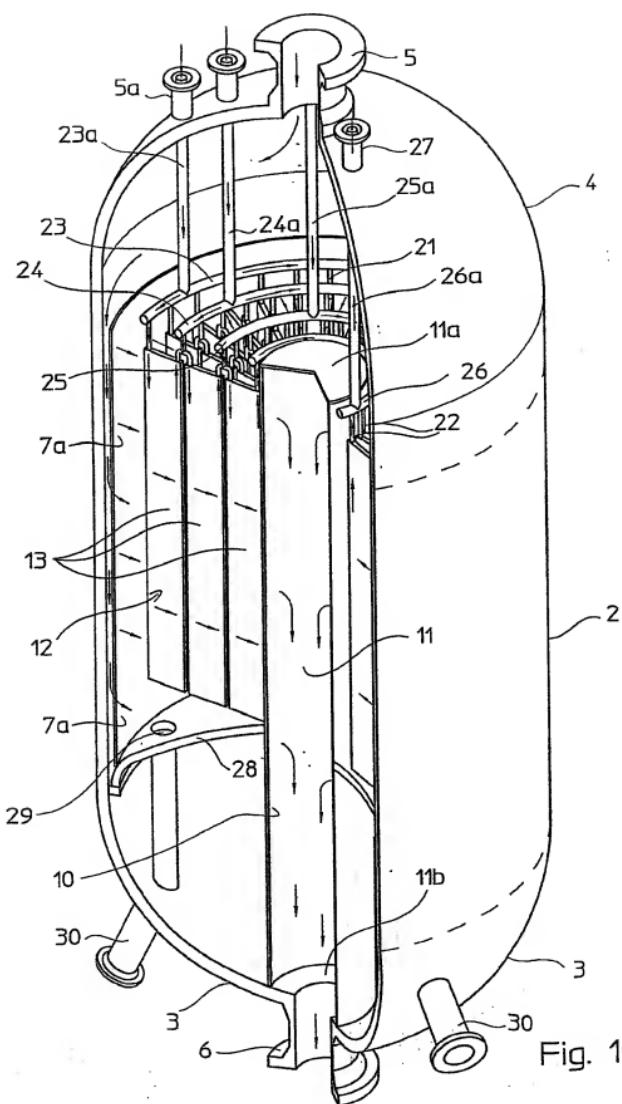


Fig. 8



## INTERNATIONAL SEARCH REPORT

International Application No  
PCT/EP 02/11024A. CLASSIFICATION OF SUBJECT MATTER  
IPC 7 B01J8/02 F28F3/14 F28D9/00

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 B01J F28F F28D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 035 867 A (DANG VU QUANG ET AL) 30 July 1991 (1991-07-30)	1-5, 9-12, 20
Y	column 1, line 7 - line 14 column 1, line 54 -column 2, line 32; figure 1	6-8, 16-19, 21, 22
X	US 3 796 547 A (MUENGER J) 12 March 1974 (1974-03-12) column 7, line 54 -column 9, line 45; figures 1-8	1-3, 9, 20
Y	US 3 595 308 A (DURBIN LEWIS H) 27 July 1971 (1971-07-27) column 3, line 16 - line 44; figures 1-3	6-8, 16-19
		-/-

 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

## \* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority, claim(s) or inventive step or which may be cited to explain an invention of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- \*Z\* document member of the same patent family

Date of the actual completion of the international search

22 January 2003

Date of mailing of the international search report

29/01/2003

## Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patenttaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax. (+31-70) 340-3016

## Authorized officer

Cubas Alcaraz, J

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 02/11024

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4 311 671 A (NOTMAN ALAN) 19 January 1982 (1982-01-19) column 8, line 58 -column 9, line 28; figure 5 _____	21,22
A	FR 2 771 802 A (DIETRICH & CIE DE) 4 June 1999 (1999-06-04) the whole document _____	1-19

## INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 02/11024

Patent document cited in search report	Publication date		Patent family member(s)		Publication date
US 5035867	A	30-07-1991	FR AT CA DE EP JP JP	2625692 A1 76330 T 1335753 A1 68901535 D1 0324670 A1 1230989 A 2745140 B2	13-07-1989 15-06-1992 06-06-1995 25-06-1992 19-07-1989 14-09-1989 28-04-1998
US 3796547	A	12-03-1974	US	3666423 A	30-05-1972
US 3595308	A	27-07-1971	NONE		
US 4311671	A	19-01-1982	AU AU CA DE EP JP NZ US	539904 B2 6217380 A 1158842 A1 3066990 D1 0026057 A1 56056225 A 194885 A 4411877 A	25-10-1984 19-03-1981 20-12-1983 19-04-1984 01-04-1981 18-05-1981 15-07-1983 25-10-1983
FR 2771802	A	04-06-1999	FR DE DE EP WO JP US	2771802 A1 69803804 D1 69803804 T2 .1036295 A1 9928691 A1 2001525532 T 6460614 B1	04-06-1999 21-03-2002 22-08-2002 20-09-2000 10-06-1999 11-12-2001 08-10-2002